CONTINUOUS RAPID CARBONIZATION OF POWDERED COAL BY ENTRAINMENT: RESPONSE SURFACE ANALYSIS OF DATA

by

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INTRODUCTION

In recent years increased interest in processes for converting coal into high-Btu pipeline gas and smokeless low-sulfur char for powerplants prompted the Morgantown Coal Research Center to study the rapid or flash carbonization of bituminous coal. The coal selected for carbonization was a strongly caking, high-volatile A bituminous from the Pittsburgh bed (34.4% V.M., 7.0% ash).

Of primary interest has been the development of a low-cost process for carbonizing high-volatile bituminous coals at high throughput rates in entrainment. Previously proposed low-temperature entrainment processes (1)[1], as well as our prior low temperature-entrainment carbonization work[2] in an externally heated 9-foot-long 4-inch-diameter isothermal reactor, yielded a high-Btu gas and a highly reactive char, along with a high yield of tar plus light oil. However, coal throughput rates were prohibitively low owing to dilute phase operation--0.35 g of coal per cu ft gas volume. Projection of such data to a commercial-scale process would result in excessively large equipment and high operating costs, although the throughput per unit cross sectional area would increase to some extent with larger carbonizers operated at higher pressures.

Dense phase entrainment, on the other hand, offered the prospect of a significant increase in coal throughput. Hence, experiments were conducted with a 4-inch-diameter by 1-foot-long carbonizer at higher temperatures with 20 times the coal concentration. The objective of this work was 3-fold: (1) to determine the effects and interactions of process variables as a guide to process feasibility; (2) to evaluate external and internal methods of applying the heat required for carbonization; and (3) to obtain data for the design of a pilot-scale carbonizer.

This paper describes these experiments, including the main steps in applying a 3-factor 5-level response surface analysis of the factorially designed test-runs, a technique that evaluates all of the significant process variables with a minimum of experimental work. Two of the 13 responses, char yield and percent volatile matter in the char, are discussed in detail.

DESCRIPTION OF EQUIPMENT

The equipment for the two test series differed in the method of heating the carbonizer and in most of the product recovery system, as shown in figures 1 and 2. The coal-feeding system was identical in both series and consisted of a vibratory screw feeder receiving coal from a pressure-equalized hopper. The feeding

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[1] Underlined numbers in parentheses refer to items in the list of references at the end of the paper.

[2] The results of these preliminary investigations will be summarized in a forthcoming U.S. BuMines Report of Investigations.

in each case was facilitated by injection of the coal into nitrogen in different dilutions with methane, before entering the flash carbonizer.

The carbonizer consisted of a 12-inch length of 4-inch-diameter, schedule 40 pipe made of type 310 stainless steel. For the external heating series, three 4-inch-long circular thermoshell heating elements were installed around the carbonizer tube. The coal particles were carbonized while being carried downward by the entraining gas. For internal heating, natural gas was burned with a slight deficiency of air in a refractory-filled combustor, the hot combustion products being injected directly into the carbonizer. The 70 percent through 200-mesh coal was further entrained in the hot gases which provided the required heat for carbonization. Internal heating changed the heat transfer from radiation controlled to turbulent convective, although some external heating was also applied to balance the heat losses from the carbonizer.

Because of the different gas flow rates for the two test series, the product recovery trains were different. In each case, the objective was complete recovery from the gas stream of all solid and liquid products. In both cases, coarser char particles fell directly into a char receiver below the carbonizer. For the externally heated unit, where the gas flow rate was lower, a baffled knockout chamber was used to remove the fine char particles. Tar and pitch were removed by two electrostatic precipitators, followed by a dry-ice trap for removal of light-oil and water, and a silica gel trap for final recovery of light-oil and water before metering and venting the gas. For the internally heated unit, which received much larger gas volumes from the combustor, the recovery train consisted of two cyclones in series for the removal of char dust, and a water scrubber followed by a steam-water scrubber for final tar, pitch, and light-oil removal.

The carbonizer was designed to rapidly heat coal particles at atmospheric pressure as they passed through the 12-inch hot zone. The feed tube was constructed to inject the powdered coal at a high velocity into the carbonization zone. After less than one second residence time, during which the particles are rapidly pyrolized and devolatilized, the char particles carried by the gas enter the recovery train.

In a typical test-run the carbonizer is preheated to the desired wall temperature. When the carbonizer temperature becomes constant at the desired level, the coal feeding is begun to start the run. During the run, char is periodically removed from the bottom lock hopper and the gas is sampled. All other products are collected after the run and a period of cooling.

EXPERIMENTAL DESIGN

To evaluate the data, the "composite factorial design" method (2) was used to obtain, in the least time, the best reliable estimates of the effects and interactions of system variables. Several of these variables, called "factors", were systematically changed and the effects in each case determined by statistical analysis.

A 3-dimensional coordinate system was assumed with three factors--coal-feed rate, reactor wall temperature, and entraining gas composition[3]--changed simultaneously while all other variables were held constant. Within this 3-dimensional

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[3]Methane is the chief component of carbonization gases and was used in order to simulate the entrainment of coal in recycle gases as would be done in pilot-scale and commercial carbonizers. Entraining gas composition is expressed in terms of methane-to-nitrogen ratios.

system, a range of values in each operating variable was chosen for examination, based on actual results. The effects and interactions of these three factors at various levels were determined on two types of responses, product-yield and quality.

The composite factorial design allowed detailed examination of the responses at 14 points plus five replications at the center of the cube, from which the entire response surface was derived. The computation of variances was based on these five replications. The three factors were varied in an established pattern as shown in table 1 and figure 3. In these illustrations, for convenience, the factors are identified as x_1 (reactor wall temperature), x_2 (coal-feed rate) and x_3 (gas composition). The levels in each factor were coded into five values: -2, -1, 0, 1, 2. The operating factors and corresponding experimental results obtained are summarized in table 2.

The main steps in response surface analysis follow:

1. Design the experiment to obtain a second degree regression equation,
2. Transform this equation into its standard or canonical form,
3. Illustrate it by means of a contour diagram or 3-dimensional model.

Assuming that a second degree equation with three factors represents the system adequately, it will have the following general form:

$$y = b_0x_0 + b_{11}x_1^2 + b_{12}x_1x_2 + b_{13}x_1x_3 + b_1x_1 + b_{22}x_2^2 + b_{23}x_2x_3 + b_2x_2 + b_{33}x_3^2 + b_3x_3 \quad (1)$$

where b_0x_0 represents the average value of all trials. This equation contains a linear term and a quadratic term for each factor and all possible 2-factor interactions. The coefficients are calculated from the data.

TABLE 1. - Composite factorial design

Factors	Level -2	Level -1	Level 0	Level 1	Level 2	Symbol
Temperature...........°F	1,500	1,600	1,700	1,800	1,900	x_1
Feed rate...........g/hr	250	500	750	1,000	1,250	x_2
Gas composition........%	100 N_2	75	50	25	0	x_3
	0 CH_4	25	50	75	100	

Note: Constants: (1) Coal type--Pittsburgh-bed hvAb, 34.4% V.M., 7.0% ash (dry basis).
(2) Coal size--70 percent through 200 mesh.
(3) Entraining gas rate--10.0 scfh.

Responses:
(1) Char yield.
(2) Char heating value.
(3) Char fuel value.
(4) Extent of devolatilization.
(5) Sulfur per Btu.
(6) Percent volatile matter.
(7) Percent sulfur.
(8) Gas yield.
(9) Gas heating value.
(10) Gas fuel value.
(11) Light-oil yield.
(12) Tar yield.
(13) Pitch yield.

TABLE 2. - Summary of data

Coded coordinates	Operating variables: Temp., °F	Feed rate, g/hr	Gas comp., % CH_4	Experimental results: External heating: % V.M.	Char yield, lb/ton	Internal heating: % V.M.	Char yield, lb/ton [1]
-1 -1 -1	1,600	500	25	23.83	1,521	15.46	1,029
-1 -1 1	1,600	500	75	25.73	1,556	15.02	1,048
-1 1 -1	1,600	1,000	25	30.11	1,732	24.81	1,253
-1 1 1	1,600	1,000	75	28.91	1,719	20.56	1,178
1 -1 -1	1,800	500	25	24.20	1,433	13.74	1,030
1 -1 1	1,800	500	75	23.20	1,454	16.01	1,074
1 1 -1	1,800	1,000	25	25.71	1,513	20.85	1,221
1 1 1	1,800	1,000	75	26.07	1,619	19.76	1,194
0 0 0	1,700	750	50	26.56	1,635	14.74	1,088
0 0 0	1,700	750	50	27.76	1,589	17.11	1,117
0 0 0	1,700	750	50	28.55	1,642	18.83	1,138
0 0 0	1,700	750	50	28.96	1,638	16.62	1,060
0 0 0	1,700	750	50	28.28	1,604	15.27	1,037
-2 0 0	1,500	750	50	31.18	1,721	23.35	1,246
2 0 0	1,900	750	50	24.65	1,468	12.05	983
0 -2 0	1,700	250	50	22.58	1,305	11.99	960
0 2 0	1,700	1,250	50	31.94	1,729	16.38	1,010
0 0 -2	1,700	750	0	29.61	1,661	16.96	1,115
0 0 2	1,700	750	100	28.90	1,593	16.32	1,000

[1] The low char yields from internal heating are attributable to some carry-over of fine dust into tar and pitch.

Since the regression equation is difficult to interpret, it is transformed into its standard or canonical form. The transformation is orthogonal and consists of translation of the original center of the design to the stationary point (where the slope with respect to all factors is zero) and rotation of the axes. Translation eliminates the linear terms; rotation causes the interactions to vanish. The transformed equation has the following general form:

$$Y - Y_s = B_{11}X_1^2 + B_{22}X_2^2 + B_{33}X_3^2 \tag{2}$$

where Y_s is the response at the stationary point and the X_i's are the coordinates with respect to the new axes after translation and rotation.

The magnitude and signs of the coefficients in the canonical equation show the nature of the response surface. If all signs are positive, the value of the response is increasing in any direction from Y_s, and Y_s has a minimum value; if all signs are negative, the value of the response is decreasing, and Y_s has a maximum value; if both positive and negative signs are present, the surface is "saddle" shaped (i.e., a ridge connecting two elevations) and there is no single maximum or minimum.

The transformation is straightforward and is based on sound mathematical procedures. The two equations, empirical and canonical, do not differ except for the location of the reference point.

All of the regression equations as well as the canonical forms have been programmed in FORTRAN IV language for the determination of the coefficients by an IBM 7040 Computer, and for plotting of the responses by an accessory CALCOMP plotter.

RESULTS

Analysis of the data by composite factorial design yields empirical equations describing the response surfaces in terms of the factors and their interactions. The coefficients of the equations indicate by their relative magnitudes the significance of the factors and the extent of their interactions as well as the curvature of the surfaces. From the equation, both linear and quadratic effects can be assessed.

The response surfaces for char yield in the external heating series are described by the regression equation:

$$y/10^3 = 1.603 - .0044x_1^2 - .0161x_1x_2 + .0131x_1x_3 - .0634x_1 - .0238x_2^2 + .0046x_2x_3 + .0917x_2 + .0037x_3^2 + .008x_3 \quad (3)$$

The coefficients of the linear terms, $0.0634x_1$ and $0.0917x_2$, are larger than any others, thus these linear effects have the greatest significance on the char yield. Statistical analysis of the data indicates that these two factors, temperature and feed-rate, are the only significant ones at the 95 percent confidence level. Based on a similar equation for the internal heating series, the char yield response is significantly dependent only upon the coal-feed rate. In absolute magnitude, these coefficients are small, indicating that the surfaces have only a slight curvature within the limits of the experiment.

The canonical equation for char yield in external heating is:

$$Y - 1{,}974 = -27.4X_1^2 - 4.7X_2^2 + 7.5X_3^2 \quad (4)$$

The form of this equation indicates that the surface is saddle-shaped since both positive and negative terms appear. The stationary point in this case is an inflection point in the surface, (i.e., neither a maximum or a minimum), and since it occurs outside the experimental limits, any description of the behavior of the surface near the point is meaningless. The saddle-shaped surface indicates only a trend in the data. Figures 4 and 5 present a comparison of the shapes of the surfaces for yields of char produced by external and internal heating, respectively.

Similarly derived from another regression equation, the 3-dimensional diagrams in figures 6 and 7 show the response surfaces for volatile matter in the char, resulting from external and internal heating, respectively. Both sets of surfaces are saddle-shaped as were those for char yield. The results of the computed data (table 3) show that the surfaces for percent volatile matter in the char depend only on coal-feed rate in the case of internal heating. With external heating, both temperature and coal-feed rate have significant effects. Table 3 also summarizes the results for the four responses discussed above.

Figure 8 shows the quantitative relationship between char yield and coal-feed rate for both the internal and external heating series. In this diagram, the wall temperature of the carbonizer and the percent volatile matter in the char are shown as parameters. The solid curves represent the char yield as a function of the coal-feed rate at the temperature levels shown. The broken lines represent similarly a functional relationship between char yield and coal-feed rate, but with percent volatile matter as a parameter at the levels shown. The diagram also shows the percent volatile matter in char as a function of the coal-feed rate at given carbonization temperatures. This follows from the fact that the char yield is a function of coal-feed rate at any carbonization temperature, as stated above.

TABLE 3. - Results of factorial analysis: yield and quality of the char produced

Response	Significant factors[1,5]	Best value of response[2]	Conditions yielding best response value[3]	Variance accounted for[4,5]	Heating Method
Char yield	Temperature, feed rate	Maximum at 15% V.M. in char	Low temperatures; high feed rates; CH_4 in gas	98.5	External
Percent volatile matter in char	Temperature, feed rate	15%	Low temperatures; low feed rates; gas composition immaterial	84.8	External
Char yield	Feed rate	Maximum at 15% V.M. in char	Low temperatures; low feed rates (250-500 g/hr); low $\%CH_4$ in gas	92.4	Internal
Percent volatile matter in char	Feed rate	15%	Surface extending from low temp.--low feed rate to high temp.--high feed rate	83.1	Internal

[1] Factors causing systematic variation in response.
[2] Desired value of response from process standpoint.
[3] Values of variables (factors) required to give the desired response.
[4] Percentage of random error accounted for in the experiment (below 80% validity of the result is poor.)
[5] At 95% confidence level.

The factorial design indicates that both temperature and coal-feed rate are significant factors at the 95 percent confidence level in the external heating series, while only the coal-feed rate is significant at the 95 percent level in internal heating. The diagrams in figure 8 reflect these conclusions. The larger effect of temperature in the external series is seen from the greater spread between the curves compared with the set of curves for internal heating. The effect of temperature is clearly present in the internal heating series, but at a much lower statistical level of confidence.

The diagrams in figure 8 show that for a given coal-feed rate, the char yield decreases with increasing carbonization temperatures, as would be expected. However, the effect of the coal-feed rate on the char yield, at higher feed rates, is contrary to what one would expect: the char yield decreases as the feed rate increases. The observed deviation in curvature can be explained by the method used to measure temperature. When the temperature of the reactor wall is measured, as in this investigation, there is a direct correlation between the true temperature of the suspended solid particles and the measured wall temperature as long as the particles are in dilute phase in the moving gas stream. However, above a certain particle concentration the correlation between wall temperature and particle temperature ceases because the particles moving downward in dense phase near the center of the carbonizer tube will not "see" as much of the source of radiant heat as the particles moving near the tube wall. In the externally heated carbonizer, this shadowing effect becomes so large that the wall temperature is no longer a true measure of the actual temperature. The same effect is confirmed by noting the percent volatile matter remaining in the char. Since the same coal is used throughout, the amount of volatile matter remaining in the char that is carbonized in dilute phase is directly related to the char yield. At high coal-feed rates in the external heating series, the curves representing the percent volatile matter in the char drop significantly with increasing coal-feed rate, also indicating that the linear relationship between wall temperature and carbonization temperature has ceased.

Differences in char yield and percent volatile matter remaining in the char for both series are evident from the diagram discussed. The upper plot shows that the minimum volatile matter left in the char from the external heating series was approximately 20 percent, or 5 percent above the 15 percent considered optimum for coal-burning powerplants. The optimum of 15% V.M. could not be attained by means of external heating over the entire range of factor levels investigated. On the other hand, the lower group of curves for the internal heating series indicate that 15 percent V.M. char can be made (with an entraining gas composition of 50 percent N_2 and 50 percent CH_4) at any temperature within the design limits of 1,500° to 1,900°F, and at coal-feed rates from 375 to 750 g/hr. The corresponding char yield was 1,075 lb per ton of coal carbonized.

A complete description of the yields and qualities of all of the products (including gas, light oil, tar and pitch) will be included in a forthcoming U.S. BuMines Report of Investigations.

CONCLUSIONS

The experimental results of the two test series show that even highly caking coals may be carbonized at a rapid rate in entrainment. The quantities and qualities of the products can be controlled within the limits of the experiment by appropriately combining the temperature, coal-feed rate and entraining gas composition.

Larger coal throughput rates, with the desired amount of volatile matter remaining in the char, were possible when the carbonizer was heated internally rather than externally because of more effective heat transfer, although at the expense of gas quality. Thus, by internal heating, optimum char quality (15% V.M.) was achieved with a throughput of 750 g/hr in a 4-inch-diameter carbonizer at 1,900°F. Comparable char quality could not be attained by external heating over the entire range of the variables investigated.

Diagrams obtained by response surface analysis of the two series of test-runs were found useful in predicting the conditions under which a product of given yield and quality can be produced. Optimization of product yields and qualities could be achieved by subsequent series of factorially designed test-runs guided by the trends indicated by the present results.

REFERENCES

1. Sebastian, J. J. S. Process for Carbonizing Coal in a Laminar Gas Stream. U.S. Patent 2,955,988, October 11, 1960.

2. Davies, O. L., Ed., The Design and Analysis of Industrial Experiments. Hafner Publishing Co., N. Y., 1963.

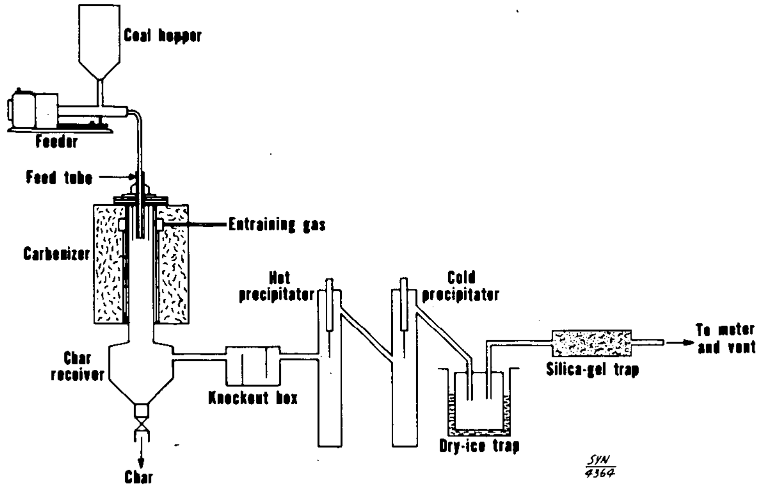


FIGURE 1. - Flash Carbonization of Powdered Coal with Externally Heated Reactor

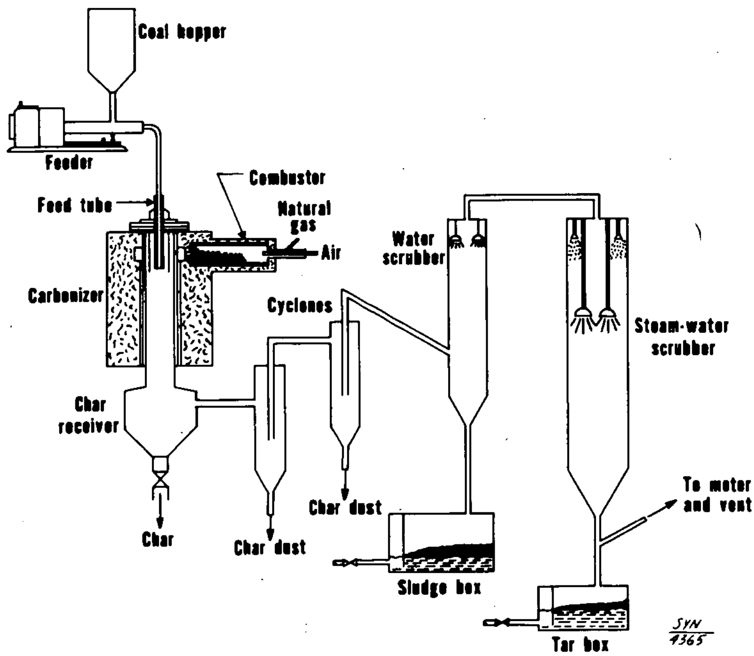
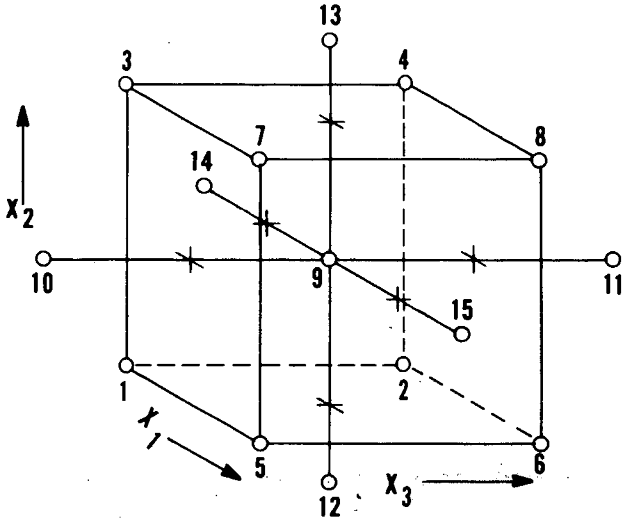


FIGURE 2. - Flash Carbonization of Powdered Coal with Internally Heated Reactor



EXPERIMENTAL POINTS IN CODED COORDINANTS

POINT	X_1	X_2	X_3
1	−1	−1	−1
2	−1	−1	1
3	−1	1	−1
4	−1	1	1
5	1	−1	−1
6	1	−1	1
7	1	1	−1
8	1	1	1
9	0	0	0
10	0	0	−2
11	0	0	2
12	0	−2	0
13	0	2	0
14	−2	0	0
15	2	0	0

FIGURE 3. - Three-Factor Composite Design

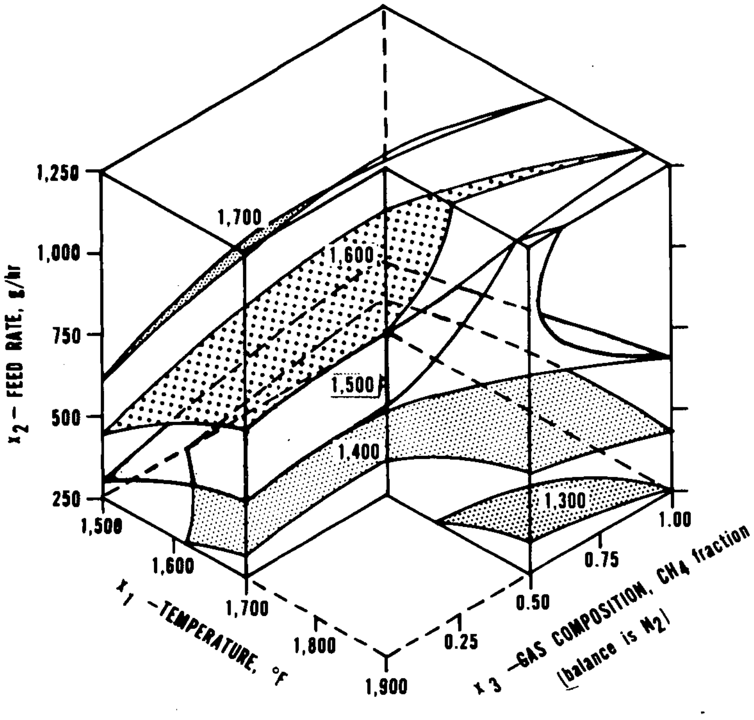


FIGURE 4. - Three-Dimensional Response Surfaces, Char Yield in lbs/ton
—Externally Heated Carbonizer—

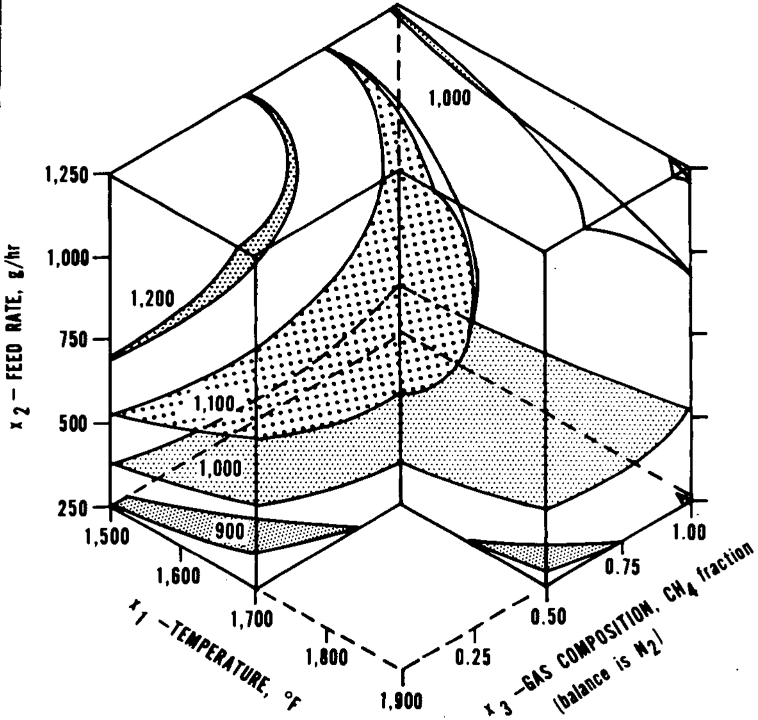


FIGURE 5. - Three-Dimensional Response Surfaces , Char Yield in lbs/ton
—Internally Heated Carbonizer—

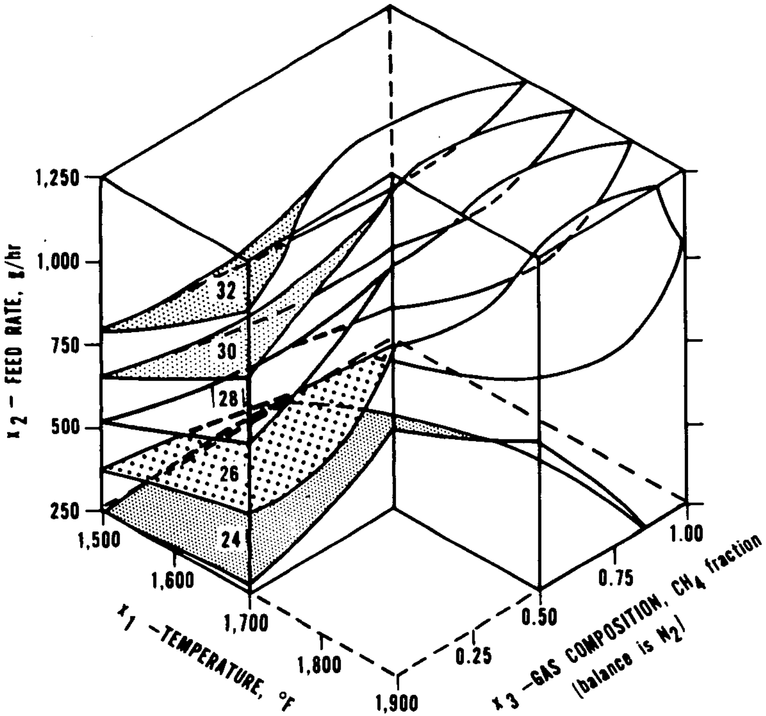


FIGURE 6. - Three-Dimensional Response Surfaces, Percent Volatile Matter in Char
—Externally Heated Carbonizer—

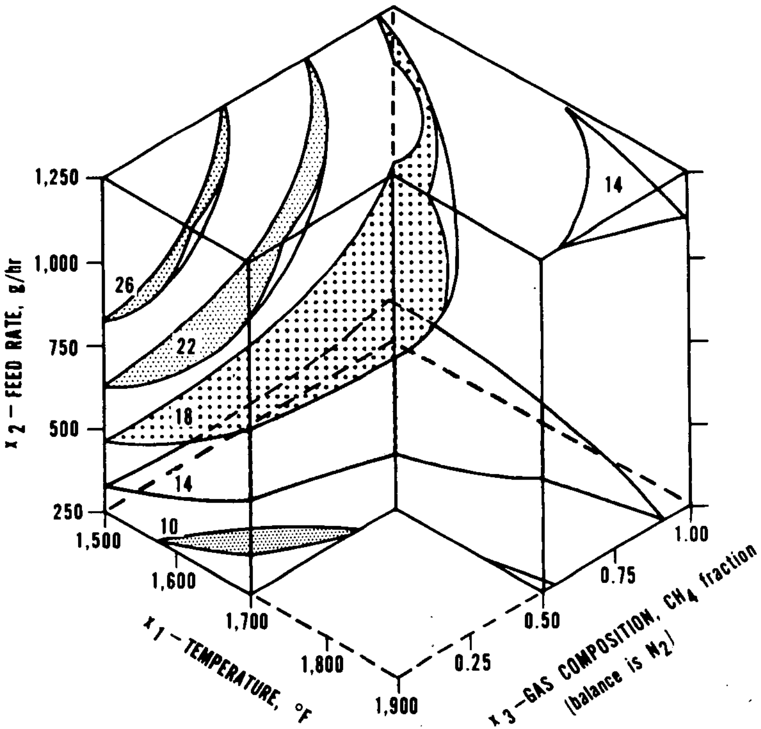


FIGURE 7. - Three-Dimensional Response Surfaces, Percent Volatile Matter in Char
—Internally Heated Carbonizer—

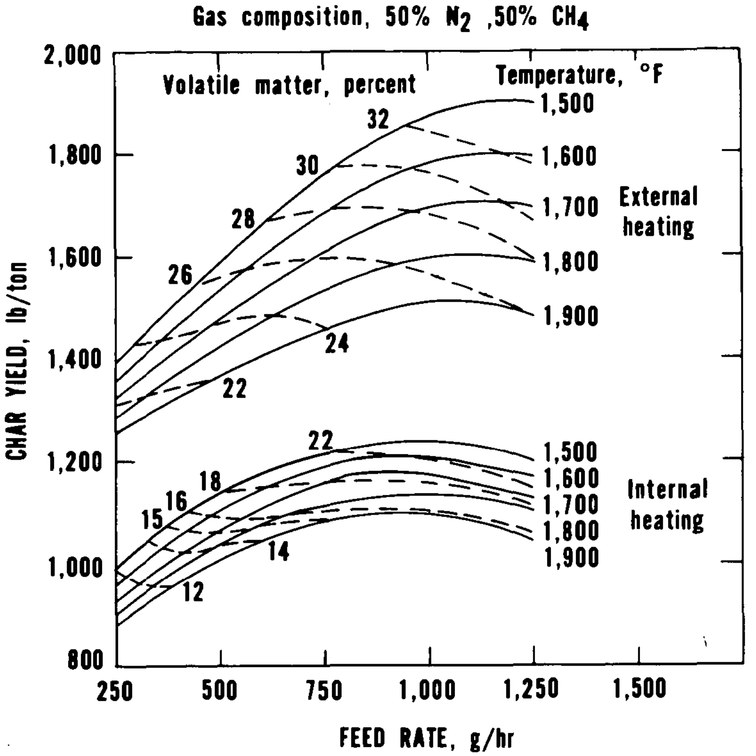


FIGURE 8. - Effect of Feed Rate and Temperature on Char Yield and Percent Volatile Matter in Char.